# Spiral correlations in frustrated one-dimensional spin-1/2 Heisenberg $J_1$-$J_2$-$J_3$ ferromagnets

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**Abstract**

We use the coupled cluster method for infinite chains complemented by exact diagonalization of finite periodic chains to discuss the influence of a third-neighbor exchange $J_3$ on the ground state of the spin-$\frac{1}{2}$ Heisenberg chain with ferromagnetic nearest-neighbor interaction $J_1$ and frustrating antiferromagnetic next-nearest-neighbor interaction $J_2$. A third-neighbor exchange $J_3$ might be relevant to describe the magnetic properties of the quasi-one-dimensional edge-shared cuprates, such as $LiVCuO_4$ or $LiCu_2O_2$. In particular, we calculate the critical point $J_2^c$ as a function of $J_3$, where the ferromagnetic ground state gives way for a ground state with incommensurate spiral correlations. For antiferromagnetic $J_3$ the ferro-spiral transition is always continuous and the critical values $J_2^c$ of the classical and the quantum model coincide. On the other hand, for ferromagnetic $J_3 \lesssim -(0.01 \ldots 0.02)|J_1|$ the critical value $J_2^c$ of the quantum model is smaller than that of the classical model. Moreover, the transition becomes discontinuous, i.e. the model exhibits a quantum tricritical point. We also calculate the height of the jump of the spiral pitch angle at the discontinuous ferro-spiral transition.

PACS codes:
75.10.Jm Quantized spin models
75.45.+j Macroscopic quantum phenomena in magnetic systems
75.10.-b General theory and models of magnetic ordering

# 1 Introduction

The recent observation of spiral (helical) magnetic ground states in several chain cuprates, such as $LiVCuO_4$, $LiCu_2O_2$, $NaCu_2O_2$, $Li_2ZrCuO_4$, and $Li_2CuO_2$ [1–11], which were identified as quasi-one-dimensional (1D) frustrated spin-1/2 magnets with ferromagnetic (FM) nearest-neighbor (NN) in-chain $J_1$ and antiferromagnetic (AFM) next-nearest-neighbor (NNN) in-chain interactions $J_2$ has stimulated intensive investigations of frustrated 1D Heisenberg ferromagnets, see, e.g., Refs. [12–21]. The 1D $J_1$-$J_2$ model considered in the most theoretical papers may serve only as the minimal model to describe the magnetic properties of these materials. Several extensions, such as exchange anisotropy [16, 21] or interchain coupling [19, 22–25] might be relevant to explain experiments. In addition to the NN and NNN exchange integrals, $J_1$ and $J_2$, also an exchange coupling to 3rd neighbors, i.e. $J_3$, or even couplings to farther distant neighbors could play a role in real materials. One mechanism to induce such exchange interactions is strong spin-phonon interaction for frustrated chains within the antiadiabatic limit [26]. Even if these additional couplings are small, their influence on the spiral ground state (GS) correlations might be noticeable. In particular, there is a significant influence of $J_3$ on the critical frustration $J_2^c$ at which the FM GS gives way for the spiral GS, see below. Except the possible relevance of the $J_1$-$J_2$-$J_3$ model for real materials the consideration of such a model is interesting in its own right as a basic model to study frustration effects in 1D quantum spin systems. Moreover, very recently it has been argued that the magnetic properties of the kagome-like mineral volborthite $Cu_3V_2O_7(OH)_2{\cdot}2H_2O$ can be described by an effective chain model with farther distant frustrating exchange couplings [27]. The corresponding general Heisenberg Hamiltonian $H$ with NN exchange $J_1$, NNN exchange $J_2$ and farther in-chain exchange interactions $J_n$ reads

$$H = \sum_n J_1 \mathbf{s}_n \mathbf{s}_{n+1} + J_2 \mathbf{s}_n \mathbf{s}_{n+2} + J_3 \mathbf{s}_n \mathbf{s}_{n+3} + \ldots \quad . \tag{1}$$

Motivated by the experiments on the edge-sharing chain cuprates we focus on the spin-1/2 $J_1$-$J_2$-$J_3$ model with FM $J_1$ and frustrating AFM $J_2 \geq 0$. To the best of our knowledge this model has been investigated so far only in an early paper of Pimpinelli et al. [12] using spin-wave theory.

Here we use the coupled cluster method (CCM) for infinite chains complemented by exact diagonalization (ED) of finite chains (periodic boundary conditions imposed) to investigate spiral GS correlations. Both methods have been successfully applied to study the spiral ordering of the $J_1$-$J_2$ model [13,19,28]. In Refs. [13,19] it was demonstrated that the CCM results are in good agreement with the DMRG data. However, in order to take into account the $J_3$ bonds properly, we go beyond the so-called SUB2-3 approximation used in Refs. [13,19] and consider an improved approximation, namely the LSUB4 approximation, see below.

The paper is organized as follows. In Sec. 2 discuss briefly the classical GS. In Sec. 3 we provide a brief illustration of the CCM and describe its application on the considered model. In Sec. 4 our results for the FM-spiral phase transition and for the pitch angle in the spiral phase are presented and discussed. In Sec. 5 we summarize our findings.

# 2 The classical model

First we discuss the GS of the classical model (spin quantum number $s \to \infty$). For the usual $J_1$-$J_2$ model, i.e. the model with $J_n = 0$, $n \geq 3$, studied in many papers the critical frustration $J_2$ is $J_2^c = |J_1|/4$. For $J_2 \geq J_2^c$ there is a spiral GS with a canting angle between NN (pitch angle) $\gamma$ given by $\cos\gamma = |J_1|/(4J_2)$. This helix interpolates between a FM chain at $0 \leq J_2 \leq J_2^c$ and two decoupled AFM chains at $J_2/|J_1| = \infty$. Noteworthy, $J_2^c$ is unaffected by quantum effects, see, e.g., Refs. [12, 13, 15, 19].

If farther AFM couplings $J_n$ $(n \geq 3)$ are relevant the destabilzation of the FM GS sets in for smaller values of $J_2$. Extending the classical model including arbitrary $J_n \neq 0$ $(n \geq 3)$ one finds for the critical NN exchange

$$J_2^c = \frac{1}{4}\left(|J_1| - \sum_{n=3}^{\infty} n^2 J_n\right). \tag{2}$$

This expression has been derived assuming a *continuous* transition between the FM and the spiral GS's. It holds for arbitrary AFM long-range couplings

$J_n$ with $n \geq 3$. In case that some of the exchange couplings are FM, i.e. $J_n < 0$ for certain $n \geq 3$, the Eq. (2) holds only if the AFM couplings dominate. Assuming $J_2 > 0$ we find as the criterion for the validity of Eq. (2)

$$J_2 \geq -\frac{1}{12}\sum_{n=3}^{\infty} n^2 \left(n^2 - 1\right) J_n, \tag{3}$$

or equivalently

$$|J_1| \geq -\frac{1}{3}\sum_{n=3}^{\infty} n^2 \left(n^2 - 4\right) J_n. \tag{4}$$

If this condition is violated, in crossing the FM-spiral phase boundary, the spiral GS "jumps" from a *finite* pitch angle $\gamma_T \neq 0$ to $\gamma = 0$ in the FM GS. For the simplest case $J_3 \neq 0$ and $J_n = 0$ $(n > 3)$ the classical model was considered by Pimpinelli et al. [12]. They found for the critical frustration $J_2$ in case of continuous transition $J_2^c = (|J_1| - 9J_3)/4$ in accordance with the general expression (2). The classical pitch angle in the spiral phase is given by $\cos\gamma = \left(-J_2 + \sqrt{J_2^2 + 3J_3(3J_3 + |J_1|)}\right)/6J_3$. According to the general Eqs. (3) and (4) for the $J_1$-$J_2$-$J_3$ model the classical FM-spiral transition is discontinuous at $J_3 < -\frac{|J_1|}{15}$ or equivalently at $J_3 < -\frac{J_2}{6}$.

For $J_3 < 0$ and $J_n = 0$, $n > 3$, the height of the jump of the pitch angle $\gamma_T$ at the transition is given by [12]

$$\cos\gamma_T = -J_1\left(2J_3 + \sqrt{4J_3^2 + 4J_3J_1}\right)^{-1} - \frac{3}{2} \tag{5}$$

or equivalently

$$\cos\gamma_T = \frac{-J_2}{4J_3} - \frac{1}{2}. \tag{6}$$

Considering other simplified classical models with a single FM long-range coupling $J_{n_0}$, i.e. a model with $J_1 < 0$, $J_2 > 0$, $J_{n_0} < 0$, $J_n = 0$ $(n > 2$ and $n \neq n_0)$, the classical discontinuous FM-spiral transitions occurs according to Eqs. (3) and (4) at $-J_{n_0} > 12J_2/\left(n_0^2\left(n_0^2 - 1\right)\right)$ (or equivalently at $-J_{n_0} > 3|J_1|/(n_0^2(n_0^2 - 4))$. In other words, even a tiny but fairly long range ferromagnetic coupling may introduce a discontinuous behavior at the critical point.

## 3 The Coupled Cluster Method (CCM)

For the sake of brevity, we will outline only some important features of the CCM which are relevant for the model under consideration. The interested reader can find more details concerning the application of the CCM on the frustrated Heisenberg magnets with non-collinear GS's in Refs. [13, 19, 29–37]. For more general aspects of the methodology of the CCM, see, e.g., Refs. [38–40].

First we mention that the CCM approach yields results in the thermodynamic limit $N \to \infty$. The starting point for a CCM calculation is the choice of a normalized reference (or model) state $|\Phi\rangle$. Related to this reference state we then define a set of mutually commuting multispin creation operators $C_I^+$, which are themselves defined over a complete set of many-body configurations $I$. For the considered frustrated spin system we choose a spiral reference state with spiral spin orientations along the chains (i.e., pictorially, $|\Phi\rangle = |\uparrow\nearrow\rightarrow\searrow\downarrow\swarrow\cdots\rangle$) characterized by a pitch angle $\gamma$, i.e. $|\Phi\rangle = |\Phi(\gamma)\rangle$. Such states include the FM state ($\gamma = 0$) as well as the Néel state ($\gamma = \pi$). Next, we perform a rotation of the local axis of the spins such that all spins in the reference state align along the negative $z$ axis. This rotation by an appropriate local angle $\delta_n$ of the spin on lattice site $i$ is equivalent to the spin-operator transformation

$$s_n^x = \cos\delta_n \hat{s}_n^x + \sin\delta_n \hat{s}_n^z \; ; \; s_n^y = \hat{s}_n^y \; ; \; s_n^z = -\sin\delta_n \hat{s}_n^x + \cos\delta_n \hat{s}_n^z, \qquad (7)$$

where $\hat{s}_n^x$, $\hat{s}_n^y$, $\hat{s}_n^z$ are the spin operators in the rotated coordinate frame. The local rotation angle $\delta_n$ is related to the pitch angle $\gamma$ of the spiral reference state by $\delta_n = n\gamma$. In this new set of local spin coordinates the reference state and the corresponding multispin creation operators $C_I^+$ are given by

$$|\hat{\Phi}\rangle = |\downarrow\downarrow\downarrow\cdots\rangle; \; C_I^+ = \hat{s}_n^+ \, , \; \hat{s}_n^+\hat{s}_m^+ \, , \; \hat{s}_n^+\hat{s}_m^+\hat{s}_k^+ \, , \; \ldots \; , \qquad (8)$$

where the indices $n, m, k, \ldots$ denote arbitrary lattice sites. In the rotated coordinate frame the Hamiltonian becomes dependent on the pitch angle $\gamma$. It reads

$$\begin{aligned} H \; = \; & \sum_{m=1}^{3} \frac{J_m}{4} \sum_n [\cos(m\gamma)+1](\hat{s}_n^+\hat{s}_{n+m}^- + \hat{s}_n^-\hat{s}_{n+m}^+) \\ + \; & [\cos(m\gamma)-1](\hat{s}_n^+\hat{s}_{n+m}^+ + \hat{s}_n^-\hat{s}_{n+m}^-) + 2\sin(m\gamma)[\hat{s}_n^+\hat{s}_{n+m}^z \\ - \; & \hat{s}_n^z\hat{s}_{n+m}^+ + \hat{s}_n^-\hat{s}_{n+m}^z - \hat{s}_n^z\hat{s}_{n+m}^-] + 4\cos(m\gamma)\hat{s}_n^z\hat{s}_{n+m}^z, \end{aligned} \qquad (9)$$

where $\hat{s}^{\pm}_{i,n} \equiv \hat{s}^{x}_{i,n} \pm \mathrm{i}\hat{s}^{y}_{i,n}$.

With the set $\{|\Phi\rangle, C_I^+\}$ the CCM parametrization of the exact ket GS eigenvector $|\Psi\rangle$ of the many-body system is given by

$$|\Psi\rangle = e^S|\Phi\rangle \ , \ S = \sum_{I\neq 0} a_I C_I^+ . \tag{10}$$

The CCM correlation operator $S$ contains the correlation coefficients $a_I$, which can be determined by the so-called set of the CCM ket-state equations

$$\langle\Phi|C_I^- e^{-S} H e^S|\Phi\rangle = 0 \ \ ; \ \forall I \neq 0, \tag{11}$$

where $C_I^- = \left(C_I^+\right)^\dagger$. Each ket-state equation belongs to a specific creation operator $C_I^+ = s_n^+,\ s_n^+ s_m^+,\ s_n^+ s_m^+ s_k^+, \cdots$, i.e. it corresponds to a specific set (configuration) of lattice sites $n, m, k, \ldots$ . By using the Schrödinger equation, $H|\Psi\rangle = E|\Psi\rangle$, we can write the GS energy as $E = \langle\Phi|e^{-S}He^S|\Phi\rangle = E(\gamma)$, which depends (in a certain CCM approximation, see below) on the pitch angle $\gamma$.
In the quantum model the pitch angle may be different from the corresponding classical value $\gamma_{\rm cl}$. Therefore, we do not choose the classical result for the pitch angle in the quantum model, rather, we consider $\gamma$ as a free parameter in the CCM calculation, which has to be determined by minimization of the CCM GS energy $E(\gamma)$, i.e. $dE/d\gamma|_{\gamma=\gamma_{qu}} = 0$.
For the many-body quantum system under consideration it is necessary to use approximation schemes in order to truncate the expansions of $S$ in Eq. (10) in a practical calculation. In Refs. [13] and [19] it has been demonstrated, that for the $J_1$-$J_2$ model the so-called SUB2-3 approximation leads to results of comparable accuracy to those obtained using the DMRG method [41]. In this approximation all configurations are included which span a range of no more than 3 contiguous sites and contain only 2 or fewer spins. Taking into account the $J_3$ bond we have to extent this approximation in order to take into account configurations including a range of 4 contiguous sites. The corresponding approximation is the so-called LSUB4 approximation, see e.g., Refs. [35, 39, 40]. Within this approximation multispin creation operators of one, two, three or four spins distributed on clusters of four contiguous lattice sites are included.

In addition, for the determination of the quantum tricitical point we have also used higher LSUB$n$ approximations, see Sec. 4. However, the

numerical complexity increases tremendously, since (i) the number of ket-state equations (11) increases exponentially with $n$, (ii) there are two free parameters $J_2$, $J_3$ which have to be varied by very small increments to find the transition points, and (iii) the determination of the quantum pitch angle $\gamma_{qu}$ itself requires the iterative minimization of the ground state energy for each set of $J_2$, $J_3$. Hence, except for the determination of the quantum tricitical point, we have restricted our CCM calculations to the CCM-LSUB4 approximation.

# 4 Results for the spin-$1/2$ quantum model

In what follows we set $J_1 = -1$ if not stated otherwise explicitly. The phase diagram of the $J_1$-$J_2$-$J_3$ chain with FM $J_1 = -1$ obtained by the CCM and by the ED is shown in Fig. 1. For the classical as well as for the quantum model the transition from the FM to the spiral GS can be second ($J_3 > J_3^T$) or first order ($J_3 < J_3^T$), i.e., the pitch angle $\gamma$ does (first order) or does not (second order) jump from $\gamma = 0$ to a finite value $\gamma = \gamma_T > 0$ at the transition. The tricritical point $T = (J_2^T, J_3^T)$, i.e. that point at the transition line where the second-order transition goes over in a first-order transition is $T_{cl} = (J_2^{T_{cl}}, J_3^{T_{cl}}) = (\frac{2}{5}, -\frac{1}{15})$ for the classical model [12], see the black square in Fig. 1. Due to quantum fluctuations this point is shifted to smaller values of $|J_3|$. The LSUB4-CCM estimate for the quantum tricritical point is $T_{qu}^{LSUB4} = (J_2^{T_{qu}^{LSUB4}}, J_3^{T_{qu}^{LSUB4}}) = (0.283, -0.013)$, see the red square in Fig. 1. This result is quite close to the spin-wave result [12] $T_{qu}^{sw} = (J_2^{T_{qu}^{sw}}, J_3^{T_{qu}^{sw}}) = (0.25, 0)$. We have determined the quantum tricritical point also using higher CCM-LSUB$n$ approximations, namely LSUB6, LSUB8 and LSUB10. The corresponding results are $(J_2^{T_{qu}^{LSUB6}}, J_3^{T_{qu}^{LSUB6}}) = (0.274, -0.011)$, $(J_2^{T_{qu}^{LSUB8}}, J_3^{T_{qu}^{LSUB8}})$ $= (0.268, -0.009)$, and $(J_2^{T_{qu}^{LSUB10}}, J_3^{T_{qu}^{LSUB10}}) = (0.266, -0.007)$. Obviously, these values are quite close to each other, and there is a slight shift towards $(J_2^{T_{qu}^{sw}}, J_3^{T_{qu}^{sw}})$ of Ref. [12] when increasing the order of CCM approximation.

The ED data for the transition line are in very good agreement with the CCM data. There is only a slight deviation for stronger FM $J_3$ bonds, i.e. for $J_3 \lesssim -0.2$. Note, however, that in a tiny interval $-0.1 \lesssim J_3 \lesssim -0.05$ for finite chains the transition from the fully polarized FM state with total spin $S = N/2$ is not directly to a singlet state with $S = 0$, rather there are

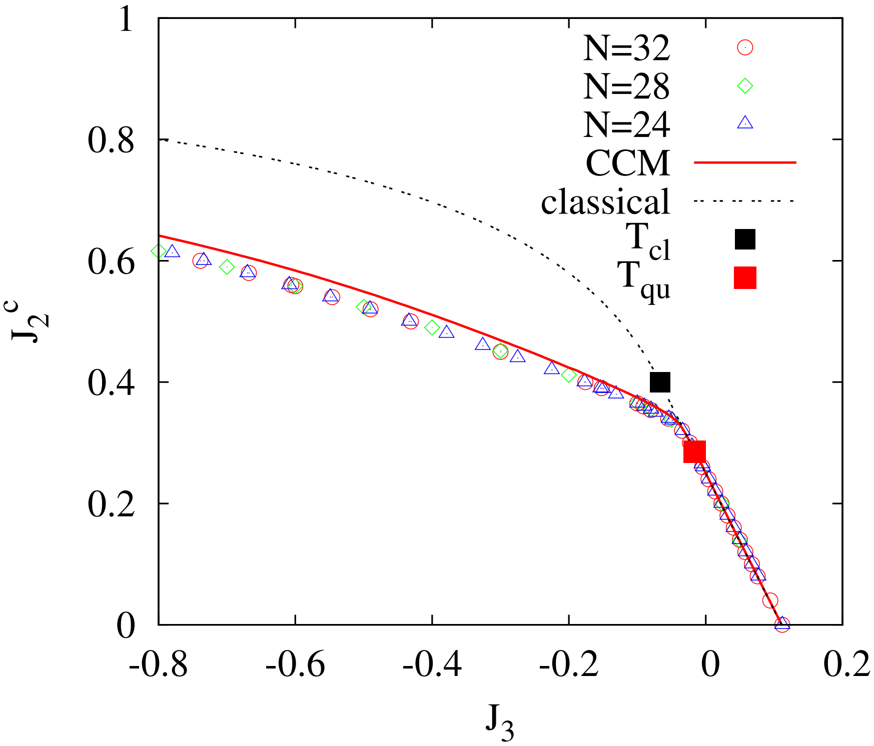


Figure 1: Phase diagram of the $J_1$-$J_2$-$J_3$ Heisenberg chain determined by CCM for $N \to \infty$ and ED for $N = 24, 28, 32$. Below the transition line the GS is the fully polarized FM state. Above the line the GS exhibits spiral correlations. The transition can be continuous (larger $J_3$) or discontinuous (smaller $J_3$), see text. Note that the classical transition line (black dashed) corresponds to the result of Pimpinelli et al. [12].

some intermediate states with $N/2 > S > 0$. To give an example, for $J_3 = -0.08$ and $N = 32$ there is a sequence of transitions from the FM state to a singlet GS via partially polarized states in the interval $0.354 < J_2 < 0.372$. The estimation of the the tricritical point by ED is $T^{ED}_{qu} = (J_2^{T^{ED}_{qu}}, J_3^{T^{ED}_{qu}}) \approx (0.28, -0.01)$, cf. Fig. 4. Note that for $J_3 > J_3^{T_{qu}}$ the classical and quantum transition lines coincide, i.e. the relation $J_2^c = (1 - 9J_3)/4$ is valid also for the quantum model. Due to the quite large prefactor $-9/4$, already a weak 3rd neighbor coupling $J_3$ has a noticeable effect on the critical $J_2^c$. For $J_3 < J_3^{T_{qu}}$ the transition point of the quantum model is shifted to smaller values of frustrating $J_2$. That is to some extent surprising, since in most of the previous studies of models exhibiting a transition between a spiral and a collinear GS in the quantum model the opposite behavior has been found, i.e., the transition to the spiral GS is shifted to higher values of frustration, see, e.g., Refs. [13, 19, 28, 29, 33, 36, 41].

To illustrate the quantum tricritical point in more detail we show in Fig. 2 the CCM results for the pitch angle versus $J_2$ for various values of $J_3$ around $J_3^{T^{LSUB4}_{qu}}$. For comparison we show also the classic pitch angle $\gamma_{cl}$. It can be clearly seen how the continuous behavior of the pitch angle $\gamma$ goes over into a discontinuous one. Interestingly, at a particular value of $J_2 = J_2^*$ the curves cross each other, i.e. the pitch angle is independent of $J_3$. For the classical model the crossing point is at $J_2^* = 1/2$ and the corresponding pitch angle is $\gamma_{cl} = \pi/3$. For the quantum model the curves do not cross exactly in a point, rather they approach each other very closely at $J_2^* = 0.335$. The pitch angle at that point is $\gamma_{qu} = 0.32\pi$. Furthermore the quantum pitch angle $\gamma_{qu}$ approaches the limiting value $\lim_{J_2\to\infty} \gamma_{qu}$ for much smaller values of $J_2$ than the classical one.

In Fig. 3 we present the height of the jump $\gamma_T$ at the transition point in dependence on $J_3$. For the classical model $\gamma_T$ is given by Eq.(5) for $J_3 < -1/15$. For the quantum model the $\gamma_T(J_3)$ curve is characterized by two nearly linear regimes, one regime (near the quantum tricritical point) with a steep increase of $\gamma_T$ and a second, almost flat one for $J_3 \lesssim -0.2$. This scenario is confirmed by the ED results for the NN spin-spin correlation function shown in Fig. 4 which may serve as a finite-chain analogue of the infinite-chain pitch angle.

Finally, let us discuss the pitch angle which appears in the limits of large $J_2$ or large $|J_3|$. This limiting value of $\gamma$ is the maximal pitch angle and it is monotonously approached from below increasing the corresponding bond

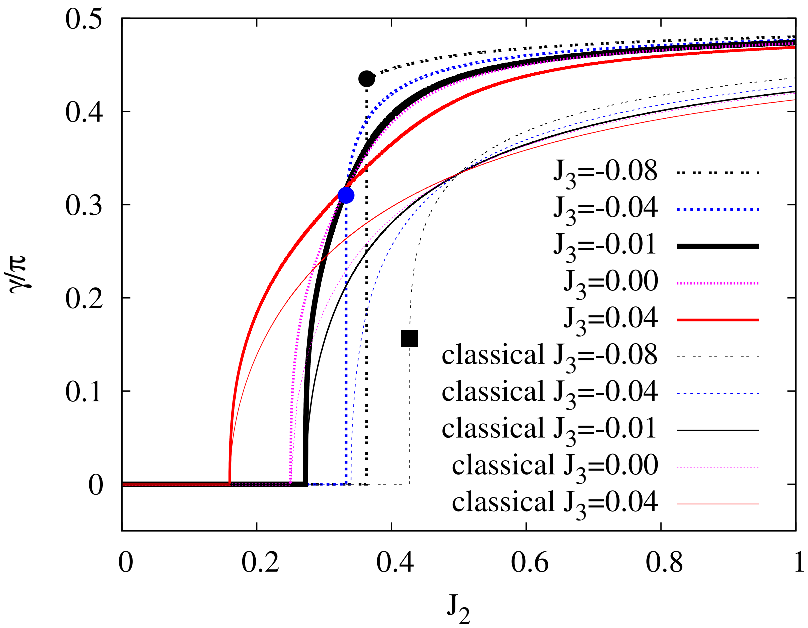


Figure 2: The CCM results for the quantum pitch angle $\gamma_{qu}$ in dependence on $J_2$ for various values of $J_3$. For comparison we also show the classical pitch angle $\gamma_{cl}$. Note that for $J_3 = -0.08$ (quantum and classical model) and for $J_3 = -0.04$ (quantum model only) the pitch angle jumps from zero to a finite value $\gamma_T$ at the transition point. These jumps are indicated by filled circles (quantum model) or a filled square (classical model).

$J_2$ or $|J_3|$ while fixing the other one. For $J_2 \to \infty$ (and finite $|J_3|$) the pitch angle approaches $\gamma = \pi/2$. In this limit the system splits into two decoupled AFM chains with coupling strength $J_2$.

For $J_3 \to \infty$ (and finite $J_2$) the pitch angle approaches $\gamma = \pi/3$, i.e. only acute pitch angles appear. For $J_3 \to -\infty$ (and finite $J_2$) the pitch angle approaches $\gamma = 2\pi/3$, i.e. by contrast to the pure $J_1$-$J_2$ model also obtuse pitch angles appear. As it is obvious from Fig. 3 pitch angles $\gamma > \pi/2$ appear already for quite moderate values of $J_3$. We mention that the above discussion is not purely academic, since at least values $J_2 > 1$ might be realized also in real materials, e.g. in $NaCu_2O_2$ [6].

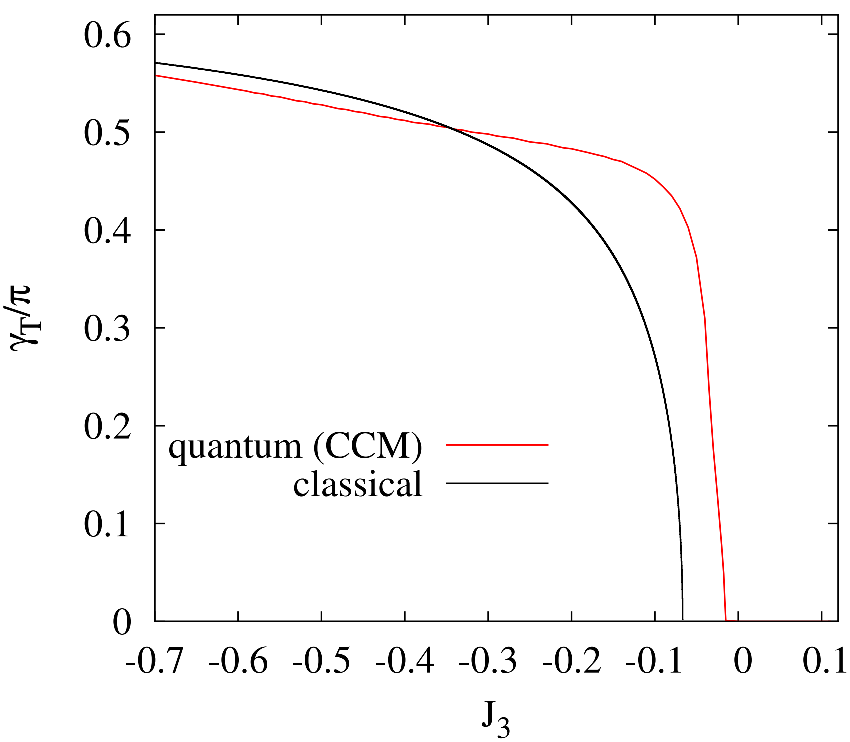


Figure 3: The height of jump of the pitch angle $\gamma_T$ at the transition point in dependence on $J_3$.

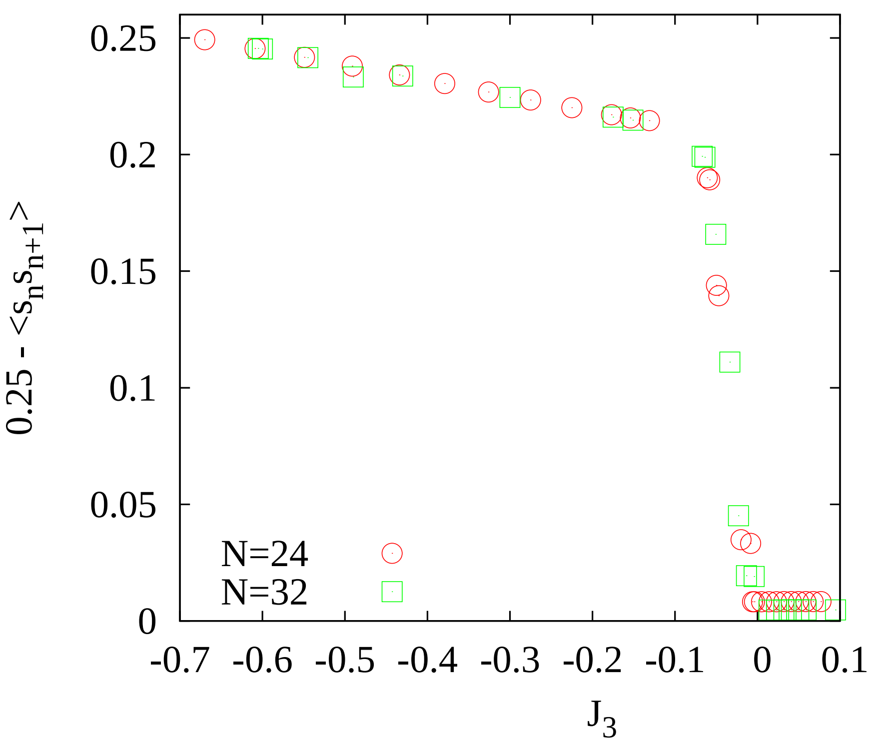


Figure 4: The jump of the nearest-neighbor spin-spin correlation function at the transition from the FM to the singlet GS, cf. Fig. 1, for finite periodic chains of length N=24 and N=32.

## 5 Summary

Using the coupled-cluster method (CCM) and the Lanczos exact diagonalization technique (ED) we have studied the influence of a third-neighbor exchange $J_3$ on the GS of the spin-$\frac{1}{2}$ Heisenberg chain with FM NN interaction $J_1$ and frustrating AFM NNN interaction $J_2$. In particular, we have analyzed the transition from the FM GS (present for dominating $J_1$) to a singlet GS with incommensurate short-range spiral correlations. The results obtained by these two different approximations agree well. Moreover, the finite-size effects inherent in the ED study appeared to be small.

We have found, that in case of an AFM coupling $J_3$ the FM-spiral transition point $J_2^c$ of the quantum model coincides with that of the classical model, and it is always continuous. However, the quantum pitch angle significantly deviates from the classical one. For a FM coupling $J_3$ quantum fluctuations shift the FM-spiral transition point $J_2^c$ to smaller values, and the transition becomes discontinuous.

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